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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re accompanying application of)
WAN S. FOONG ET AL)
Filed February 27, 2004)
FILTER) February 27, 2004
_____)

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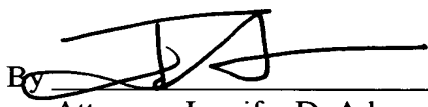
CLAIM TO PRIORITY

Applicant reaffirms the claim for the benefit of filing date of the following foreign patent application referred to in Applicant's Declaration:

Singapore Application Serial No. 200300833-1 - filed February 28, 2004

A copy of the application certified by the Singapore Registry of Patents is enclosed.

Respectfully submitted,
WAN S. FOONG ET AL

By 
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Registration No. 47,379
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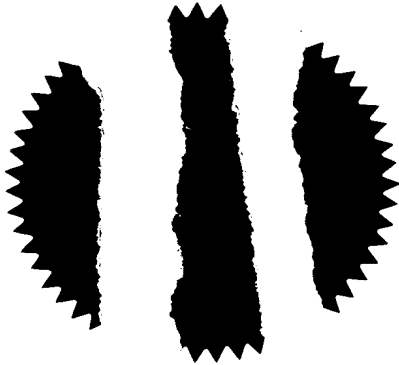
This is to certify that the annexed is a true copy of following application as filed with the Registry.

Date of Filing : 28 FEBRUARY 2003

Application Number : 200300833-1

Applicant(s) /
Proprietor(s) of Patent : SHELL INTERNATIONALE RESEARCH
MAATSCHAPPIJ B.V.

Title of Invention : PROCESS




SHARMAINE WU (Ms)
Assistant Registrar
for REGISTRAR OF PATENTS
SINGAPORE

5 FEBRUARY 2004

INTELLECTUAL PROPERTY OFFICE OF SINGAPORE
REQUEST FOR THE GRANT OF A PATENT UNDER
SECTION 25

101101

* denotes mandatory fields

1. YOUR REFERENCE*

CG/CWY/PAT/8112539/SG

2. TITLE OF
INVENTION*

PROCESS

3. DETAILS OF APPLICANT(S)* (see note 3)

Number of applicant(s)

1

(A) Name

SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ B.V.

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State

Country

NL

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For corporate applicant

☐

For individual applicant

State of incorporation

State of residency

Country of incorporation

NL

Country of residency

☐

For others (please specify in the box provided below)

(B) Name

Address

State

Country

☐ For corporate applicant

☐ For individual applicant

State of incorporation

State of residency

Country of incorporation

Country of residency

☐ For others (please specify in the box provided below)

(C) Name

Address

State

Country

☐ For corporate applicant

☐ For individual applicant

State of incorporation

State of residency

Country of incorporation

Country of residency

☐ For others (please specify in the box provided below)

☐

Further applicants are to be indicated on continuation sheet 1

4. DECLARATION OF PRIORITY (see note 5)

A. Country/country designated

File number

Filing Date

DD MM YYYY

B. Country/country designated

File number

Filing Date

DD MM YYYY

☐

Further details are to be indicated on continuation sheet 6

5. INVENTOR(S)* (see note 6)

A. The applicant(s) is/are the sole/joint inventor(s)

Yes

☐

No

☒

B. A statement on Patents Form 8 ~~is~~ will be furnished

Yes

☒

No

☐

6. CLAIMING AN EARLIER FILING DATE UNDER (see note 7)

☐ section 20(3)

☐ section 26(6)

☐ section 47(4)

Patent application number

DD MM YYYY

Filing Date

Please mark with a cross in the relevant checkbox provided below
(Note: Only one checkbox may be crossed.)

☐ Proceedings under rule 27(1)(a)

DD MM YYYY

Date on which the earlier application was amended

☐ Proceedings under rule 27(1)(b)

7. SECTION 14(4)(C) REQUIREMENTS (see note 8)

Invention has been displayed at an international exhibition. Yes

☐

No

☒

8. SECTION 114 REQUIREMENTS (see note 9)

The invention relates to and/or used a micro-organism deposited for the purposes of disclosure in accordance with section 114 with a depository authority under the Budapest Treaty.

Yes

☐

No

☒

9. CHECKLIST*

(A) The application consists of the following number of sheets

i.	Request	<input type="text" value="05"/>	Sheets
ii.	Description	<input type="text" value="14"/>	Sheets
iii.	Claim(s)	<input type="text" value="02"/>	Sheets
iv.	Drawing(s)	<input type="text" value="-"/>	Sheets
v.	Abstract (Note: The figure of the drawing, if any, should accompany the abstract)	<input type="text" value="01"/>	Sheets
Total number of sheets		<input type="text" value="22"/>	Sheets

(B) The application as filed is accompanied by:

☐ Priority document(s)

☐ Translation of priority document(s)

☐

Statement of inventorship
& right to grant

☐

International exhibition certificate

10. DETAILS OF AGENT (see notes 10, 11 and 12)

Name

Firm

DREW & NAPIER LLC
20 RAFFLES PLACE #17-00, OCEAN TOWERS, SINGAPORE 048620

11. ADDRESS FOR SERVICE IN SINGAPORE* (see note 10)

Block/Hse No.

--

Level No.

--

Unit No./PO Box

152

Street Name

ROBINSON ROAD

Building Name

--

Postal Code

900302

12. NAME, SIGNATURE AND DECLARATION (WHERE APPROPRIATE) OF APPLICANT OR AGENT* (see note 12)
(Note: Please cross the box below where appropriate.)

☒ X

I, the undersigned, do hereby declare that I have been duly authorised to act as representative, for the purposes of this application, on behalf of the applicant(s) named in paragraph 3 herein.



Name and Signature

DD MM YYYY

28 02 2003

NOTES:

1. This form when completed, should be brought or sent to the Registry of Patents together with the rest of the application. Please note that the filing fee should be furnished within the period prescribed.
2. The relevant checkboxes as indicated in bold should be marked with a cross where applicable.
3. Enter the name and address of each applicant in the spaces provided in paragraph 3.
Where the applicant is an individual
- Names of individuals should be indicated in full and the surname or family name should be underlined.
- The address of each individual should also be furnished in the space provided.
- The checkbox for "For individual applicant" should be marked with a cross.

Where the applicant is a body corporate
- Bodies corporate should be designated by their corporate name and country of incorporation and, where appropriate, the state of incorporation within that country should be entered where provided.
- The address of the body corporate should also be furnished in the space provided.
- The checkbox for "For corporate applicant" should be marked with a cross.

Where the applicant is a partnership
- The details of all partners must be provided. The name of each partner should be indicated in full and the surname or family name should be underlined.
- The address of each partner should also be furnished in the space provided.
- The checkbox for "For others" should be marked with a cross and the name and address of the partnership should be indicated in the box provided.
4. In the field for "Country", please refer to the standard list of country codes made available by the Registry of Patents and enter the country code corresponding to the country in question.
5. The declaration of priority in paragraph 4 should state the date of the previous filing, the country in which it was made, and indicate the file number, if available. Where the application relied upon in an International Application or a regional patent application e.g. European patent application, one of the countries designated in that application [being one falling under section 17 of the Patents Act] should be identified and the country should be entered in the space provided.
6. Where the applicant or applicants is/are the sole inventor or the joint inventors, paragraph 5 should be completed by marking with a cross the 'YES' checkbox in the declaration (A) and the 'NO' checkbox in the alternative statement (B). Where this is not the case, the 'NO' checkbox in declaration (A) should be marked with a cross and a statement will be required to be filed on Patents Form 8.
7. When an application is made by virtue of section 20(3), 26(6) or 47(4), the appropriate section should be identified in paragraph 6 and the number of the earlier application or any patent granted thereon identified. Applicants proceeding under section 26(6) should identify which provision in rule 27 they are proceeding under. If the applicants are proceeding under rule 27(1)(a), they should also indicate the date on which the earlier application was amended.
8. Where the applicant wishes an earlier disclosure of the invention by him at an International Exhibition to be disregarded in accordance with section 14(4)(c), then the 'YES' checkbox at paragraph 7 should be marked with a cross. Otherwise, the 'NO' checkbox should be marked with a cross.
9. Where in disclosing the invention the application refers to one or more micro-organisms deposited with a depository authority under the Budapest Treaty, then the 'YES' checkbox at paragraph 8 should be marked with a cross. Otherwise, the 'NO' checkbox should be marked with a cross. Attention is also drawn to the Fourth Schedule of the Patents Rules.
10. Where an agent is appointed, the fields for "DETAILS OF AGENT" and "ADDRESS FOR SERVICE IN SINGAPORE" should be completed and they should be the same as those found in the corresponding Patents Form 41. In the event where no agent is appointed, the field for "ADDRESS FOR SERVICE IN SINGAPORE" should be completed, leaving the field for "DETAILS OF AGENT" blank.
11. In the event where an individual is appointed as an agent, the sub-field "Name" under "DETAILS OF AGENT" must be completed by entering the full name of the individual. The sub-field "Firm" may be left blank. In the event where a partnership/body corporate is appointed as an agent, the sub-field "Firm" under "DETAILS OF AGENT" must be completed by entering the name of the partnership/body corporate. The sub-field "Name" may be left blank.
12. Attention is drawn to sections 104 and 105 of the Patents Act, rules 90 and 105 of the Patents Rules, and the Patents (Patent Agents) Rules 2001.
13. Applicants resident in Singapore are reminded that if the Registry of Patents considers that an application contains information the publication of which might be prejudicial to the defence of Singapore or the safety of the public, it may prohibit or restrict its publication or communication. Any person resident in Singapore and wishing to apply for patent protection in other countries must first obtain permission from the Singapore Registry of Patents unless they have already applied for a patent for the same invention in Singapore. In the latter case, no application should be made overseas until at least 2 months after the application has been filed in Singapore, and unless no directions had been issued under section 33 by the Registrar or such directions have been revoked. Attention is drawn to sections 33 and 34 of the Patents Act.
14. If the space provided in the patents form is not enough, the additional information should be entered in the relevant continuation sheet. Please note that the continuation sheets need not be filed with the Registry of Patents if they are not used.



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PROCESS

The present invention relates to a process for preparing organic hydroperoxides and to processes in which such organic hydroperoxides are used, such as the preparation of oxirane compounds and preparation of alkenyl aryl compound.

Background of the invention

Processes for preparing propylene oxide employing organic hydroperoxides, are well known in the art. As described in US-A-5,883,268, such process conventionally comprises peroxidation of ethylbenzene, followed by contacting the peroxidation reaction product with aqueous base in amount sufficient to neutralize acidic components thereof and separating the resulting mixture into an aqueous stream and a deacidified organic stream. The base contaminated, deacidified hydroperoxide stream is washed with water and the resulting mixture separated into an organics contaminated water phase and an organic phase having a reduced alkali metal content.

Surprisingly, it was found that the amount of contaminants in the organic hydroperoxide finally obtained, can be reduced further in a simple and effective way.

Summary of the invention

The present invention relates to a process for preparing organic hydroperoxides, which process comprises:

(a) oxidation of an organic compound to obtain reaction product containing organic hydroperoxide,

(b) treating at least part of the organic hydroperoxide containing reaction product with a basic aqueous solution and separating hydrocarbonaceous phase containing organic hydroperoxide from basic aqueous phase,

(c) washing with water at least part of the separated hydrocarbonaceous phase containing organic hydroperoxide and separating hydrocarbonaceous phase containing organic hydroperoxide from aqueous phase, and

(d) optionally repeating step (c) one or more times, in which process solid particles are removed from the reaction product containing organic hydroperoxide and/or basic aqueous solution before use in step (b).

Preferably, the solid particles are removed by filtering at least part of the reaction product containing organic hydroperoxide and/or at least part of the basic aqueous solution before use in step (b).

The gist of the present invention resides in the fact that solid particles are removed from at least part of one or more of the feed streams before these feed streams are used in step (b). The feed streams which are subjected to process step (b) are the organic hydroperoxide containing reaction product obtained in step (a) and the basic aqueous solution with which the organic hydroperoxide containing reaction product is treated.

It was observed that in conventional operation, an interface emulsion layer or rag could form upon washing the hydrocarbonaceous phase containing organic hydroperoxide. Without wishing to be bound to any theory, it is thought that such layer causes an increased amount of basic aqueous solution in the organic phase. The presence of the basic aqueous solution in the organic phase tends to cause problems in the further processing of the organic hydroperoxide due to the presence of basic

compounds, more specifically compounds such as sodium hydroxide and calcium hydroxide.

Organic hydroperoxides are useful in a range of processes. One of these processes is the reaction of organic hydroperoxide with olefin in order to obtain oxirane compounds. In such process, the organic compound usually is an alkylaryl compound, and the process further comprises:

- (e) contacting at least part of the hydrocarbonaceous phase containing alkylaryl hydroperoxide obtained in step (c) and/or (d) with olefin and catalyst to obtain alkylaryl hydroxide and oxirane compounds, and
- (f) separating at least part of the oxirane compound from the alkylaryl hydroxide.

The alkylaryl hydroxide obtained in step (f) can be used in a wide range of processes. Such process is preparing an alkenyl aryl compound by dehydrating the alkylaryl hydroxide. Another process is hydrogenating the alkylaryl hydroxide to obtain an alkylaryl compound. If the process according to the present invention is to be used for dehydrating the alkylaryl hydroxide, the process suitably comprises further:

- (g) converting at least part of the alkylaryl hydroxide obtained in step (f).

Preferably, step (g) comprises either dehydration or hydrogenolysis of the reaction product. Hydrogenolysis is the reaction of the alkylaryl hydroxide with hydrogen, preferably in the presence of catalyst. Dehydration will generally produce an alkenyl aryl compound and water, while hydrogenolysis will generally produce alkylaryl compound. Preferably, the hydrogenolysis will produce the alkylaryl compound used as starting compound.

Detailed description of the invention

Although the organic compound used in the process of the present invention can in principle be any compound, organic compounds which are most frequently used are alkylaryl compounds. Alkylaryl compounds which are most frequently used are benzene compounds containing at least 1 alkyl substituent which alkyl substituent contains of from 1 to 10 carbon atoms, preferably of from 2 to 8 carbon atoms. Preferably, the benzene compound contains on average of from 1 to 2 constituents. The alkylaryl compounds most frequently encountered are ethylbenzene, cumene and di(iso-propyl)benzene.

The oxidation of the organic compound can be carried out by any suitable process known in the art. The oxidation can be carried out in the liquid phase in the presence of a diluent. This diluent is preferably a compound which is liquid under the reaction conditions and does not react with the starting materials and product obtained. However, the diluent can also be a compound necessarily present during the reaction. For example, if the alkylaryl compound is ethylbenzene the diluent can be ethylbenzene as well.

Besides the desired organic hydroperoxide, a wide range of contaminants are created during the oxidation of organic compounds. Although most of these are present in small amounts, the presence of the organic acids especially has been found to sometimes cause problems in the further use of the organic hydroperoxides. As described in US-A-5,883,268, a method of reducing the amount of contaminants is contacting the reaction product containing organic hydroperoxide with an aqueous alkali solution. However, contact with the aqueous alkali solution introduces a certain amount of alkali metal into the organic hydroperoxide containing reaction product. The amount of organic acids is decreased by an alkali

wash but the amount of alkali metal contaminants is increased.

In the process of the present invention, the organic hydroperoxide containing reaction product is contacted with a basic aqueous solution, more specifically a basic aqueous solution containing one or more alkali metal compounds. Suitable alkali sources for use in the aqueous alkali solution include alkali metal hydroxides, alkali metal carbonates and alkali metal hydrogen carbonates. Examples of these compounds are NaOH, KOH, Na₂CO₃, K₂CO₃, NaHCO₃ and KHCO₃. In view of their easy availability, it is preferred to use NaOH and/or Na₂CO₃.

The basic aqueous solution preferably contains fresh basic aqueous solution, recycled basic aqueous solution and optionally additional water. The recycled basic aqueous solution has been obtained from step (b).

The exact conditions under which step (b) is carried out, strongly depend on the further circumstances. Preferably, step (b) is carried out at a temperature of between 0 °C and 150 °C, more preferably of between 20 °C and 100 °C.

In step (b), the hydrocarbonaceous phase is subsequently separated from the aqueous phase. A preferred method comprises allowing the hydrocarbonaceous phase and aqueous phase to settle in a settling vessel and subsequently separating a hydrocarbonaceous phase from an aqueous phase. Preferably, the hydrocarbonaceous phase containing organic hydroperoxide is subsequently sent to a coalescer where further aqueous phase is removed. Preferably, the separation is carried out at a temperature of between 0 °C and 150 °C, more preferably of between 20 °C and 100 °C.

In conventional operation, an interface emulsion layer or rag was sometimes observed upon washing the hydrocarbonaceous phase containing organic hydroperoxide.

Surprisingly, it has now been found that such rag formation could be prevented by removing solid particles from one or more of the feed streams used in step (b). Without wishing to be bound to any theory, it is thought that the rag formation was due to the presence of solid particles, such as small, insoluble particles of metal compounds such as iron. Such metal compounds can be formed in the corrosion of metal surfaces. The organic hydroperoxide containing reaction product could pick up such metal compounds during oxidation. Furthermore, the stream containing the organic compound can already contain such solid particles before the oxidation. The basic aqueous solution could pick up such metal compounds from recycled basic aqueous solution and/or from waste water which is used in the preparation of the basic aqueous solution. Both feed streams can pick up metal compounds during storage. Solid particles can be removed in different ways. Solid particles can be removed in any way known to someone skilled in the art. Suitable methods comprise treating at least part of one or more of the feed streams used in step (b) with an ion exchange resin, with an adsorbent and/or filtering at least part of these feed streams. It was found that filtering was the preferred method of removing solid particles.

The temperature and pressure at which the filtering can be carried out, are well known to someone skilled in the art and depend on the compounds present.

The filter which is preferably used for filtering the feed streams of step (b) has openings of 50 micrometres or less, preferably 30 micrometres or less, more preferably 20 micrometres or less.

The filter can be made of any material which is known to be suitable by someone skilled in the art. Filters made of polypropylene and cellulose were found to perform well. It is well known that the filters will

slowly plug which is shown by an increased pressure drop over the filter. When the pressure drop becomes too high, the filter can be taken out of operation, cleaned and be returned as will be well known to someone skilled in the art. Alternatively, the filter can be cleaned by feeding a clean liquid such as cumene or ethylbenzene in the reverse direction of the normal flow, so-called back-flushing. The latter has the advantage that the filter does not need to be removed.

As mentioned above, each of the feed streams which is used in step (b) can contain metal compounds which need to be removed. The amount of metal compounds which is incorporated in the organic hydroperoxide containing reaction product produced in step (a) depends on the amount of metal compounds present in the organic compound subjected to step (a) and on the exact processing conditions in step (a). The amount and kind of metal compounds present in step (a) will determine whether solid particles need to be removed from the product of step (a), or part of it.

The basic aqueous solution used in step (b) can pick up metal compounds from various sources. The amount of metal compounds present in each source, will determine when solid particles are preferably removed from the basic aqueous phase.

In order to further improve the separation of aqueous phase and hydrocarbonaceous phase in steps (b), (c) and/or optional step (d), additional compounds can be present. Examples of such additional compounds are so-called emulsion breakers or de-hazers such as aliphatic or cyclic amines.

In step (c), at least part of the separated hydrocarbonaceous phase obtained is washed with water. The washing can be carried out with clean water and/or with waste water.

In the present description of the invention, the expression water is used to indicate both clean water and waste water which can contain contaminants. If clean water is to be used, this is mentioned separately. The washing with water of steps (c) and (d) can be carried out with clean water and/or waste water.

It was recently found that the use of waste water, optionally in combination with clean water, has many advantages. This has been described extensively in not-prepublished patent application PCT/EP02/10519 (our TS 1068). Therefore, the washing with water of steps (c) and/or (d) preferably is carried out with waste water optionally in combination with clean water.

The waste water can be added to separated hydrocarbonaceous phase at any stage. A preferred, specific embodiment comprises adding waste water or aqueous solution containing waste water to a coalescer.

Preferably, the water used in step (c) and/or (d) comprises both waste water previously used in washing a hydrocarbonaceous phase containing organic hydroperoxide and a different kind of waste water.

The waste water previously used in washing a hydrocarbonaceous phase containing organic hydroperoxide, preferably is a waste water obtained by contacting a hydrocarbonaceous phase containing organic hydroperoxide with an aqueous phase, preferably clean water, and subsequently separating the aqueous phase from the hydrocarbonaceous phase. The aqueous phase so obtained is preferably used as waste water without further treatment. Most preferably, the waste water obtained in this way is used in combination with a different kind of waste water.

The washing of the hydrocarbonaceous phase is preferably carried out by contacting the hydrocarbonaceous phase countercurrently with water. Countercurrent operation is considered to comprise

contacting with relatively clean water hydrocarbonaceous phase which has already been washed once or more, while contacting hydrocarbonaceous phase which has not yet been washed, with aqueous phase which already has been in contact with hydrocarbonaceous phase.

The source of waste water is in principle irrelevant to the present process. However, it is preferred that the waste water is obtained in a process step related to the present process as this reduces the risk that the compounds present in the hydrocarbonaceous phase react with those present in the aqueous solution. Furthermore, it is preferred not to introduce new components into the process. It is surprising that the use of waste water gives good results as the aim of the previous process steps was to remove organic acids which were formed as by-products in the oxidation of step (a). It has now been found that waste water can be used in the aqueous wash of step (c) and/or (d), giving good results without negative impact on a subsequent catalyst such as an epoxidation catalyst such as described in EP-A-345856.

Waste water which has been found especially suitable for use in aqueous solutions for the present invention, is waste water which is acidic. Preferably, the acidic waste water comprises one or more organic acids. Organic acids have been found to be generally compatible with the compounds further used in the present process. It has been found especially preferred if the acid which is present is an organic acid comprising of from 1 to 20 carbon atoms. Preferred organic acids to be present in the waste water include hydrocarbyl carboxylic acids having in total from 1 to 10 carbon atoms. Especially preferred acids are formic acid, acetic acid, propionic acid and butyric acid. It has been found that formic acid is especially suitable as formic acid was observed to

give only limited decomposition of the organic hydroperoxide.

The concentration of acid in the aqueous solution preferably is from 0.0001 to 5 %wt, based on total amount of aqueous solution, more preferably from 0.001 to 2 %wt, most preferably from 0.001 to 1 %wt.

Preferably, the water for use in steps (c) and/or (d) consists of waste water optionally in combination with clean water and has a pH of from 2 to 7, preferably of from 3 to less than 7, more preferably of from 3.5 to 6.5.

Waste water streams can be used as such. However, in some cases it might be advantageous to concentrate the waste water stream before use in the process according to the present invention.

Dependent on the amount of contaminants present in the hydrocarbonaceous phase containing organic peroxide, the washing with water is either carried out once or a number of times. Preferably, the washing is carried out of from 1 to 3 times.

In optional process step (e), at least part of the hydrocarbonaceous phase containing organic hydroperoxide obtained in steps (c) and/or (d) is contacted with olefin, preferably propene, in the presence of a catalyst to obtain alkylaryl hydroxide and oxirane compounds. A catalyst which can suitably be used in such process comprises titanium on silica and/or silicate. A preferred catalyst is described in EP-A-345856. The reaction generally proceeds at moderate temperatures and pressures, in particular at temperatures in the range of from 0 to 200 °C, preferably in the range from 25 to 200 °C. The precise pressure is not critical as long as it suffices to maintain the reaction mixture as a liquid or as a mixture of vapour and liquid. Atmospheric

pressure may be satisfactory. In general, pressures can be in the range of from 1 to 100×10^5 N/m².

The oxirane compounds can be separated from the reaction product containing alkylaryl hydroxide in any way known to be suitable to someone skilled in the art. The liquid reaction product may be worked up by fractional distillation, selective extraction and/or filtration. The solvent, the catalyst and any unreacted olefin or alkylaryl hydroperoxide may be recycled for further utilization.

The alkylaryl hydroxide obtained in the process can be dehydrated in the presence of a catalyst to obtain styrene and water. Process which can be used for this step have been described in WO 99/42425 and WO 99/42426. However, any suitable process known to someone skilled in the art can in principle be used.

The present invention is further illustrated by the following examples.

Comparative Example 1

In a reactor, air was blown through ethylbenzene. The product obtained contained ethylbenzene hydroperoxide. This product was contacted with a solution containing 0.5 %wt NaOH in water and mixed at a temperature of 60 °C. The weight ratio of product containing ethylbenzene hydroperoxide to NaOH containing solution was 4.5:1 (wt:wt). The neutralized mixture obtained was sent to a settling vessel where a neutralized hydrocarbonaceous phase containing ethylbenzene hydroperoxide was separated from an aqueous phase.

The neutralized hydrocarbonaceous phase containing ethylbenzene hydroperoxide was sent to a coalescer where further aqueous phase was removed. The neutralized hydrocarbonaceous phase containing ethylbenzene

hydroperoxide, was washed by mixing the neutralized ethylbenzene hydroperoxide solution from the coalescer with an aqueous solution, separating the mixture obtained in a settling vessel into an aqueous phase and a hydrocarbonaceous phase, subsequently separating the hydrocarbonaceous phase obtained from the settling vessel with the help of a first coalescer, and separating the hydrocarbonaceous phase obtained in the first coalescer with the help of a second coalescer. Each of these steps is described in more detail herein below. The hydrocarbonaceous phase obtained in the second coalescer contains ethylbenzene hydroperoxide, ethyl benzene, water and contaminants. This hydrocarbonaceous phase is distilled. The distillate contains ethyl benzene, water and contaminants. This distillate was phase separated in a vessel to obtain a hydrocarbonaceous phase containing ethyl benzene and contaminants, and an aqueous phase containing water and contaminants. The latter had a pH of 3 and was used as wastewater for use in the aqueous solution for washing the neutralized hydrocarbonaceous phase.

The neutralized ethylbenzene hydroperoxide solution was mixed with an aqueous solution in a ratio of 4.5:1 (wt:wt). The aqueous solution comprised 85 %wt of water which is being recycled in this process step to which is added 1.3 %wt of clean water and 13.7 %wt of wastewater which had been used in washing a hydrocarbonaceous phase containing organic hydroperoxide.

The mixture which was obtained, was sent to a settling vessel where a hydrocarbonaceous phase was separated from an aqueous phase.

NaOH was added to the aqueous phase obtained, which NaOH containing aqueous phase was for use in the neutralization of the hydrocarbonaceous phase containing ethylbenzene hydroperoxide.

The hydrocarbonaceous phase obtained in the settler, was sent to a first coalescer where were added 1.1 %wt (based on total hydrocarbonaceous phase) of the distillate aqueous phase containing water and contaminants described above, and 1.7 %wt (based on total hydrocarbonaceous phase) of clean water. An aqueous phase and a hydrocarbonaceous phase were obtained in the first coalescer. The hydrocarbonaceous phase from the first coalescer was sent to the second coalescer where further 1.4 %wt (based on total hydrocarbonaceous phase) of clean water were added.

It was found that the hydrocarbonaceous phase obtained from the second coalescer, contained about 1 ppm of sodium.

Example 1

Comparative Example 1 was repeated except that the NaOH containing aqueous phase which was recycled to the neutralization step was filtered with the help of a Whatman polypropylene filter having openings of at most 0.4 micrometers before being used again in the neutralization step.

It was found that the hydrocarbonaceous phase obtained from the second coalescer, contained substantially less than 1 ppm of sodium. Additionally, it was found that the pressure over the filter gradually increased from $0.05 \times 10^5 \text{ N/m}^2$ to $1 \times 10^5 \text{ N/m}^2$ in the course of 3 weeks. This indicates that solids are being separated off.

Example 2

Waste water was obtained in the dehydration of 1-phenyl ethanol to styrene. The waste water obtained was distilled whereby the distillate obtained contains water and organic compounds. Organic phase was separated off from the distillate in a settler. The aqueous phase was sent from the settler to a coalescer. The aqueous phase

obtained in the coalescer contained 10 ppm of solids of which 2 ppm was iron. To this aqueous phase was added 20 %wt of NaOH. Thus obtained NaOH solution was filtered with the help of a polypropylene filter having openings of different maximum sizes. The filtrate was contacted with a solution of ethylbenzene hydroperoxide in ethylbenzene at 70 °C for several hours. In a comparative example, the NaOH solution had not been filtered before use. The following results were obtained.

filter size (micrometre)	none	40	20	10	6
rag formation	strong	slight	none	none	none

C L A I M S

1. Process for preparing organic hydroperoxides, which process comprises:

- (a) oxidation of an organic compound to obtain reaction product containing organic hydroperoxide,
- (b) treating at least part of the organic hydroperoxide containing reaction product with a basic aqueous solution and separating hydrocarbonaceous phase containing organic hydroperoxide from basic aqueous phase,
- (c) washing with water at least part of the separated hydrocarbonaceous phase containing organic hydroperoxide and separating hydrocarbonaceous phase containing organic hydroperoxide from aqueous phase, and
- (d) optionally repeating step (c) one or more times, in which process solid particles are removed from the reaction product containing organic hydroperoxide and/or basic aqueous solution before use in step (b).

2. Process according to claim 1, in which process solid particles are removed by filtering at least part of the reaction product containing organic hydroperoxide and/or at least part of basic aqueous solution before use in step (b).

3. Process for preparing oxirane compounds, which process comprises preparing an alkylaryl hydroperoxide with the help of the process according to claim 1 or 2 in which the organic compound is an alkylaryl compound, and which process further comprises:

- (e) contacting at least part of the hydrocarbonaceous phase containing alkylaryl hydroperoxide obtained in step (c) and/or (d) with olefin and catalyst to obtain alkylaryl hydroxide and oxirane compounds, and
- (f) separating at least part of the oxirane compound from the alkylaryl hydroxide.

4. Process for preparing an alkenyl aryl compound, which process comprises preparing an alkylaryl hydroxide with the help of the process according to claim 3, which process further comprises:

(g) converting at least part of the alkylaryl hydroxide obtained in step (f).

5. Process according to any one of claims 2 to 4, in which process the filtering is carried out with the help of a filter having openings of 50 micrometres or less.

6. Process according to any one of claims 2 to 5, in which process at least part of the basic aqueous solution which has been separated off in step (b) is filtered and subsequently recycled to step (b).

7. Process according to any one of claims 1 to 6, in which process the basic aqueous solution contains fresh basic aqueous solution, recycled basic aqueous solution and optionally additional water.

8. Process according to any one of claims 1 to 7, in which process step (c) and/or (d) at least part of the separated hydrocarbonaceous phase containing organic hydroperoxide is washed with waste water optionally in combination with clean water.

A B S T R A C T

PROCESS



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Process for preparing organic hydroperoxides, which process comprises (a) oxidation of an organic compound to obtain reaction product containing organic hydroperoxide, (b) treating at least part of the organic hydroperoxide containing reaction product with a basic aqueous solution and separating hydrocarbonaceous phase containing organic hydroperoxide from basic aqueous phase, (c) washing with water at least part of the separated hydrocarbonaceous phase containing organic hydroperoxide and separating hydrocarbonaceous phase containing organic hydroperoxide from aqueous phase, and (d) optionally repeating step (c) one or more times, in which process solid particles are removed from the reaction product containing organic hydroperoxide and/or basic aqueous solution before use in step (b), preferably by filtering, and a process for preparing oxirane compounds in which such organic hydroperoxide is used.